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The ability of crystal violet to mode-lock the rhodamine 6G laser is reported. The relaxation time of crystal violet is solvent dependent. The manner in which the mode-locking is affected by varying this relaxation time is described. A 2 mm path of a  $\times 10^{-5}$  M solution of crystal violet in glycerol having a relaxation time of 100 ps, gave the best quality mode-locking for the 48 cm long  $\times$  8 mm diameter dye laser used.

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# CRYSTAL VIOLET AS A MODE-LOCKER OF THE RHODAMINE 6G DYE LASER

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## Abstract

The ability of crystal violet to mode-lock the rhodamine 6G laser is reported. The relaxation time of crystal violet is solvent dependent. The manner in which the mode-locking is affected by varying this relaxation time is described.

### 1. Introduction

The fluorescence yield and lifetime of crystal violet (CV) in solution have been shown to be strongly dependent upon solvent viscosity [1,2]. Work in this laboratory has shown that the rate of return of ground state absorption in crystal violet after excitation at 530 nm is also viscosity dependent and in the picosecond regime [3]. Through selection of the proper solvent, the ground state repopulation time can be varied from tens to hundreds of picoseconds. The absorption spectrum of crystal violet in ethanol has its maximum at 590 nm ( $\epsilon \sim 10^5 \text{ M}^{-1}\text{-cm}^{-1}$ ) and is similar to that of DODCI (3,3' diethyl oxadicarbocyanine iodide), which is commonly used to mode-lock the rhodamine 6G dye laser (figure 1). This similarity, combined with the fast relaxation of CV, suggested its application as a mode-locker and our results support this idea.

### 2. Experimental

The mode-locking ability of CV was tested on a CANDELA ED-625 coaxial flashlamp-pumped dye laser, modified to reduce the cross-section of the lasing medium from 25 to 8 mm. This modification improved the quality of the mode-locking, presumably by discriminating against off-axis modes. Distilled water was circulated between the flashlamp and dye cell (39 cm long) to provide cooling and to increase optical coupling. The 48 cm long cavity was formed by an 80% reflector ( $R = \infty$ ) and a grating. According to New [4] a cavity of this length is near optimum, since the round trip cavity time is of the order of the rhodamine 6G lifetime of 4.8 nsec. The optically flat and anti-reflection-coated dye cell windows were slightly misaligned to prevent reflections along the lasing axis. None of the components in the cavity was set at Brewster's angle.

(Greek epsilon  $\epsilon$ )  
(infinity  $\infty$ )

The CV mode-locking solution was placed in direct contact with the output mirror by using a specially-constructed cell having a 2 mm path length [5,6]. Initially the path length was 6 mm, but 2 mm improved the quality of mode-locking. The window on the cell was intentionally slightly misaligned.

Laser output was monitored using a fast photodiode (HP-5082-4203)-fast oscilloscope (Tektronix 7904) combination. The lasing wavelength was determined using a Gaertner dual prism spectrometer and was recorded on Polaroid 4" by 5" type 57 film.

### 3. Results

Rhodamine 6G (Eastman Kodak) was dissolved in 95% ethanol at a concentration of  $5 \times 10^{-5}$  M. "Best" mode-locking (reproducible complete modulation) occurred with a CV in reagent glycerol concentration of  $\sim 3 \times 10^{-5}$  M. Changes in the concentration above or below this value reduced the wavelength range over which good mode-locking took place. CV worked well over the range 590 - 610 nm. Outside this range the quality of the trains was reduced. Shot to shot reproducibility was good over the range cited. The pulse trains (figure 2a) consisted of 90 - 100 pulses separated by the round trip cavity time of  $\sim 5$  nsec (figure 2b).

Qualitatively speaking, it appears, for our laser at least, that CV is a better mode-locker than DODCI. The pulse trains produced by DODCI (in ethanol) do not show 100% modulation until at least 100 nsec into the train, whereas CV (in glycerol) gives total modulation over the entire train. The induction period for mode-locking by DODCI probably arises from the need to form a photoisomer [7,8]. Our observation of such an induction period in the 590 - 610 nm region provides direct support for this hypothesis and further suggests that mode-locking, even within the normal ground-state absorption region of DODCI, requires the presence of the much shorter-lived photoisomer.

We also made a qualitative study of the effect on the mode-locking of varying the ground state repopulation (GSR) time  $\tau$  of CV. Using the solvents listed in Table 1, mixtures of CV were prepared having concentrations of  $3 \times 10^{-5}$  M. Of the four solutions, reagent glycerol proved most satisfactory. Mode-locking was also obtained using CV in glucose-glycerol, but good pulse trains were observed less often. For this latter solution it was more critical to operate the laser as close as possible to threshold. The CV-glycerol-water solution also showed

(Greek tau  $\tau$ )

100% modulation, but the degree of reproducibility was less than for reagent glycerol solutions. In the case of the ethanol solution ( $\tau < 10$  psec), we saw no modulation that could be attributed to crystal violet. To be sure, there was slight modulation of the output, but it was present in the absence of CV and hence is probably self-modulation by the lasing dye.

#### 4. Discussion

Studies by Bradley et al [6] have demonstrated that optimum generation of picosecond pulses results when the saturable absorber is placed in a short path length cell in contact with one of the cavity mirrors. Both these observations and our own results can be understood in terms of the following model.

The shortest duration pulses and best mode-locking will be obtained when the cell path length is small enough to allow the incident and reflected pulse to overlap over the entire cell thickness. If the cell is too thick to satisfy this condition and the absorber relaxes very rapidly, the reflected pulse must rebleach the absorber on its return journey through the cell. For cells that are thin, as defined by the above criterion, and for thicker cells also, provided the absorber relaxation is slow compared to the pulse transit time, the reflected pulse finds the absorber still bleached and passes through essentially unattenuated. The need in some circumstances (thick cells, fast absorber relaxation) to bleach the absorber twice in succession may account for the results reported by Bradley et al [6] that the quality of mode-locking improved when the cell path length was reduced. A cell thin enough to permit overlap of incident and reflected pulse is required only when the absorber relaxation time is very short. For absorbers of longer relaxation time,  $\tau$ , thicker cells will still allow a substantially unattenuated return journey provided that  $\tau \gg 2 n l/c$ , where  $l$  is the cell thickness and  $n$  the refractive index of the absorbing medium. Of course, if  $\tau$  is made too long, the absorber does not discriminate against undesirable satellite pulses following the main bleaching pulse. This too should cause a reduction in mode-locking quality, as evidenced by our results with the 300 psec relaxation time absorber.

In our experiments the quality of mode-locking improved as the cell thickness was reduced from 6 mm to 2 mm, with the absorber relaxation time maintained constant. In contrast, for the 2 mm cell, the mode-locking deteriorated when the absorber relaxation time was reduced from

100 psec to 30 psec and disappeared completely when the ethanol solution ( $\tau < 10$  psec) was used. Both these observations support the above model. Further support is provided by the results of Ippen and Shank, who were unable to achieve stable mode-locking using malachite green, which has a relaxation time of 2.1 psec [9]. To sum up, we can say that, when the relaxation time of the absorber is of the order of a few times ( $2 \text{ nL}/\text{c}$ ), further shortening of  $\tau$  or increase of cell thickness  $l$  both lead to a progressive deterioration in the quality of mode-locking.

### 5. Conclusions

We have shown that crystal violet is capable of efficient mode-locking of the rhodamine 6G laser. Crystal violet with a GSR time of 100 psec created completely mode-locked pulse trains with good reproducibility. GSR times greater or shorter by a factor of three reduced the reproducibility.

The 100 psec GSR time we report as being optimum may be peculiar to the laser system we used. However, the utility of CV as a mode-locker has been demonstrated, and because its relaxation time can be easily varied, by appropriate choice of solvent, it should prove useful in a variety of laser systems.

### Acknowledgements:

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Figure Captions

Figure 1. Emission spectrum of rhodamine 6G (in ethanol) in relation to the absorption spectra of crystal violet (in glycerol) and DODCI (in ethanol).

Figure 2. (a) Oscillogram of mode-locked pulse train. Duration of train is approximately 500 nsec. (b) Tracing of an oscillogram showing individual pulses. Resolution is limited by resolution of detection system.

Table 1

Solvent (% wt)	Viscosity (poise)	GSR lifetime, $\tau$ (psec)
ethanol	0.011	10
83.5% reagent glycerol in water	0.67	32
reagent glycerol	4.45	100
30% glucose in reagent glycerol	86	300

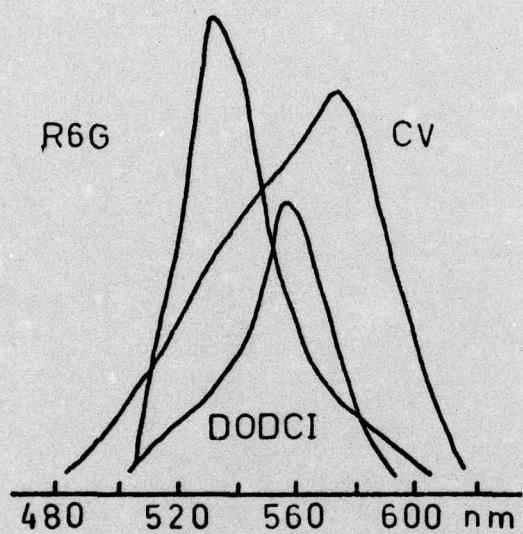
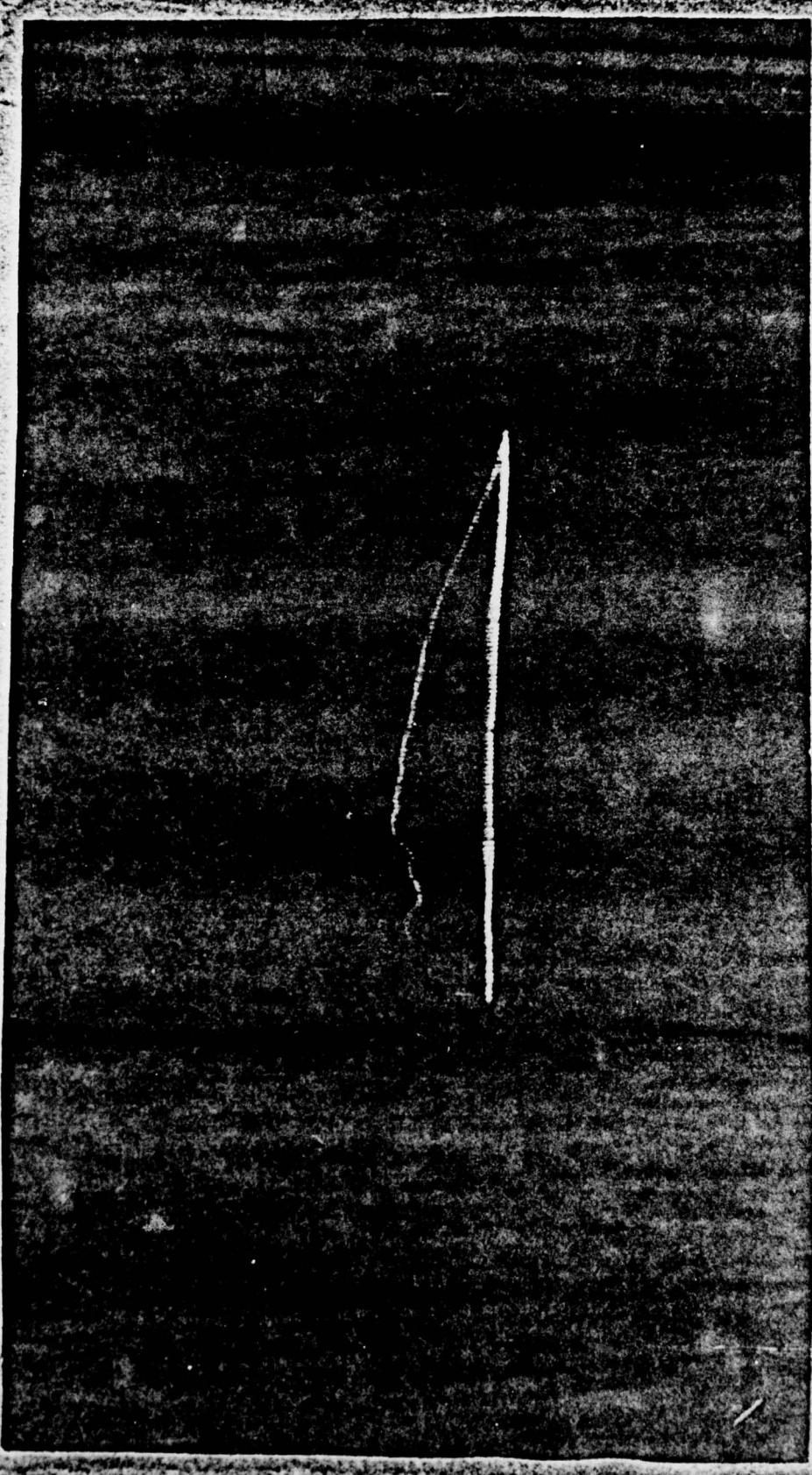


Figure 1

FIGURE 2a



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Figure 2b

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